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LASER-INDUCED DAMAGE IN OPTICAL MATERIALS

Concetto R. Giuliano

Hughes Research Laboratories

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I. INTRODUCTION AND SUMMARY

In this report we present the results of our research on fundamental problems of laser induced damage in optical materials. The activity during this period was centered at two optical wavelengths 0.694 and 1.06 μm using ruby and Nd:YAG lasers with well characterized and monitored spatial and temporal properties. The damage investigations of proustite have continued at 1.06 μm; some new results have been obtained with regard to obtaining a better surface finish but no substantial increase in damage threshold has been achieved. The phenomenon of ion beam polishing has been explored in a preliminary way for $LiNbO_3$ on a number of different samples; the treatments have not been successful so far, but the damage appears to be inclusion limited and not an intrinsic property of the host material.

The fundamentals of surface damage have been explored further using the Q-switched ruby laser. In those experiments the temporal and spatial properties of backscattered and specularly reflected light have been studied and compared with transmitted light temporal profile. It is found that an appreciable amount of light is scattered out of the beam in the back direction at the time of damage. Some preliminary evidence for precatastrophic damage is presented in the form of temporal irregularities in the backscattered pulse in the absence of any other evidence for damage. The possibility that signs of damage can be seen before catastrophic failure occurs is an exciting one and opens up an important area for investigation.

II. DAMAGE EXPERIMENTS AT 1.06 MICRONS

A. Damage Experiments in Proustite (Ag_3AsS_3)

During this period we have continued to study damage in proustite at 1.06 μm using the high power Nd:YAG laser described in the last report. The pulse duration of this laser (FWHM) is in the range 16.5 to 18.5 nsec. For the damage experiments reported earlier the laser output was focused on the proustite entrance surface using a 10.6 cm focal length lens. More recently it has been convenient to employ a shorter focal length lens (3.3 cm) for the proustite damage experiments. The experimental setup is essentially the same as that employed earlier and described in Semiannual Technical Report No. 1. The gaussian spot size measurements at the location of the sample surface were carried out in the same manner as described previously.

The main purpose of the proustite experiments during this period was to explore the possibility of obtaining a better surface finish and hence possibly a higher damage threshold. A number of different polishing methods were tried both with respect to polishing compound and lapping surface; a variety of results were obtained, mostly negative. For example, Syton, a Monsanto product which is an alkaline suspension of SiO_2 in water, was tried as a polishing compound. This material has been found to be an excellent polishing compound for a variety of different materials from metals to refractory oxides and glasses but on proustite the result was a blackening and severe etching of the surface presumably via a chemical reaction with the hydroxide ions.

Procedures which did result in some improvement in surface quality were the use of 0.05 μm fumed silica in water using a medium pitch lap, as well as 0.05 μm γ-alumina in water on a pitch lap. The main different between these polishing methods and the one used earlier is that a paraffin lap was used in the early polishing methods. Although some qualitative improvement was achieved in surface finish (e.g., lower density of scratches) there was not an appreciable increase in damage threshold.

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Table I shows results obtained for different polishing conditions on the same sample.

Several features should be brought up in discussing the results presented in Table I. First, the reason for the difference between the results obtained for the two spot sizes is not completely clear. The data were taken at different times and the surface condition of the sample was not necessarily the same although it was prepared in the same manner. It is possible that the average value of damage threshold might be higher for a smaller spot size because the chance of encountering a low threshold region (assuming a certain distribution of absorbing inclusions) on each shot is less in general for a small spot than for a larger one. But one would expect that even though the average value might be higher, the range of values should overlap somewhat, especially on the low side (i.e., one should occasionally encounter low threshold regions with the small spot size as well). Of course, the number of thresholds measured is not large enough in these cases to preclude the possibility that this spot size effect is a reflection of the distribution of inclusions and/or surface defects.

TABLE I

Damage Thresholds for Proustite Sample With Different Polishing Conditions (17.5 nsec pulse)

Polishing Conditions	Damage Threshold,[a] J/cm^2 (average value)	Range	Number of Thresholds Measured	Beam radius,[b] μm
γ-Al_2O_3 on paraffin	0.715	0.54 to 1.0	17	86
γ-Al_2O_3 on paraffin	1.31	1.17 to 1.49	11	45
Fumed silica on pitch	1.5[illegible]	1.29 to 2.02	14	45
Fumed silica on pitch (fresh polish)	1.71	1.04 to 3.33	13	45

[a] These values are given as the total energy divided by the beam area defined as πa^2 where a is the $1/e$ radius for the electric field. The energy densities on-axis are twice as large as the values quoted in the table.

[b] Defined at the $1/e$ points for the electric field (the $1/e^2$ points for the intensity) for a gaussian beam.

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Of more significance, however, is the last entry labeled "fresh polish." In most instances, as discovered earlier, the threshold measurements on proustite are carried out on samples that have been recently polished, usually on the same day. This practice was undertaken because it was found that the surface was seen to accumulate a cloudy appearance within a few hours of polishing when viewed with appropriate illumination under magnification. Typically the damage thresholds would begin to drift downward as the cloudiness would develop; after one or two days of standing in air the threshold would level off to a value sometimes as much as a factor of two lower than those obtained in the first few hours of measurement.

This type of behavior was most striking in the series of experiments that resulted in the last entry in Table I (fresh polish). This phenomenon is illustrated in Fig. 1 which shows the threshold results as obtained sequentially for a series of spots on the sample surface each separated by 0.5 mm. The gradual dropoff referred to above can be seen in the figure.

The threshold value reported in Table I is an average of all the thresholds measured. However, it can be seen (Fig. 1) that the first few points measured result in lifting the average considerably. The total sequence of measurements illustrated in Fig. 1 represent about 100 shots of the laser and required most of a full day to carry out. This kind of behavior illustrates the difficulty one faces in obtaining meaningful damage threshold data for proustite.

B. <u>Proustite Damage on a Sample From a Different Source</u>

In addition to the series of experiments just described we had the opportunity to measure damage threshold on a proustite sample obtained from a different source than previous samples. Earlier proustite crystals were either grown at HRL or obtained from the Royal Radar Establishment (RRE). Samples from these sources had about the same damage threshold (within an average of ~50%) for similar conditions of pulse duration and focusing. Recently we obtained a slice from a proustite boule grown by the British Drug House (BDH), formerly a supplier of raw material for proustite crystal growth.

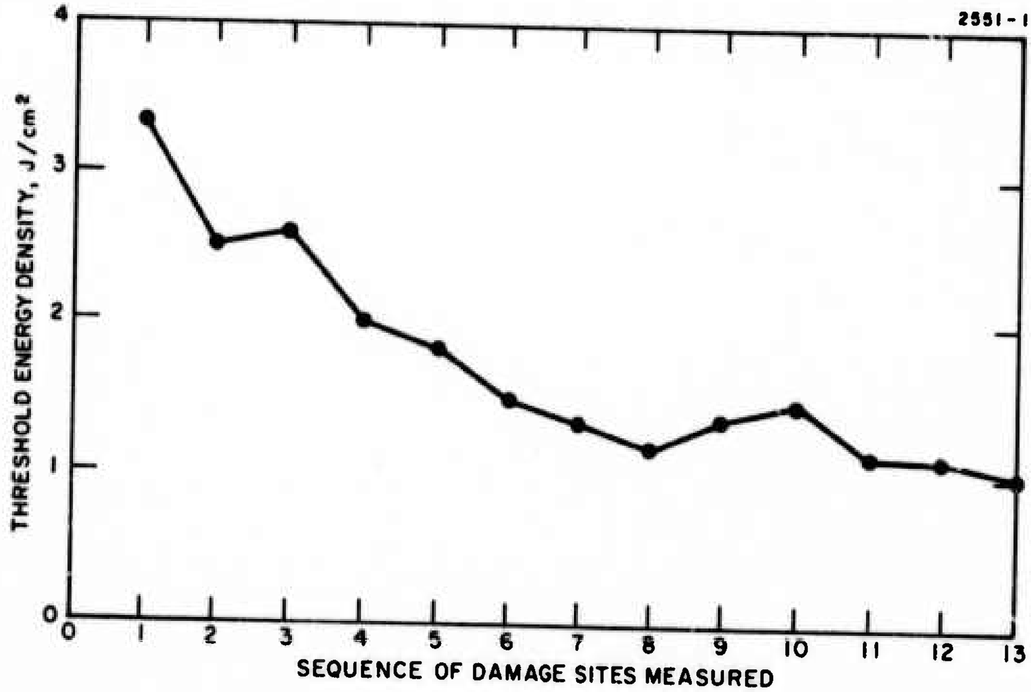

Fig. 1. Values of damage thresholds obtained in sequence on a freshly polished proustite surface.

The sample was polished by the technique described (fumed silica, pitch lap) and a series of surface damage threshold measurements was carried out.

As a comparison to the entries for the sample in Table I (an RRE sample), the BDH sample was damaged under the same focusing conditions as the latter entries (45 μm spot radius). The damage threshold energy density was 1.57 J/cm^2, an average of 27 different sites with values ranging from 1.1 to 2.0 J/cm^2.

One feature that distinguished the BDH sample from previously studied proustite samples is the morphology for surface damage formed near threshold. As illustrated in Semiannual Technical Report No. 1, the pulsed damage near threshold is characterized by a number of tiny molten areas clustered around surface scratches. Besides, the damage formed near threshold is rarely accompanied by the occurrence of a spark; surface sparks are usually seen on proustite if the incident energy is appreciably above threshold. For the BDH sample, however, the damage in all but 2 of the 27 cases observed consisted of a single surface pit and was accompanied by a spark.

The reason for this is not completely clear but we believe it is an indication of a more uniform surface quality on this sample. The damage thresholds measured are not appreciably different from those measured under similar conditions for the RRE sample, but the fact that the morphology consisted of a single damage crater rather than a cluster of molten globules indicates that if the damage is limited by inclusions or other absorbing sites in the BDH sample they are more uniformly distributed than they are in the RRE samples studied.

C. Chemical Changes Accompanying Damage in Proustite

During this period we have explored the damage sites in proustite with respect to the possibility of chemical changes. As mentioned in the last report, the catastrophic surface damage caused by cw illumination is accompanied by a tiny plume of yellow smoke (presumably sulfur) emanating from the damage site.

This suggestion of chemical decomposition during laser damage led us to confirm it by other means and to determine whether similar decomposition might accompany other types of proustite surface damage (e.g., pulsed) as well.

To explore the surface composition we used an electron excited x-ray microprobe apparatus. This phenomenon employs a focused electron beam on the surface that locally excites atoms found in the beam. These excited atoms emit characteristic x rays that are measured in an x-ray spectrometer. The apparatus used has a scanning electron beam capability as well, and the different regions being probed can be displayed as on an electron microscope.

Results of a microprobe analysis are shown in Fig. 2. Here we see an electron micrograph of a damage site caused by cw illumination. Accompanying the micrograph is a series of dot patterns taken over the same region of the surface as seen in the micrograph. The brightness of the dot pattern in a given region is a measure of the amount of the element in question found in that region.

From these pictures we see (not surprisingly) that there is a definite chemical change accompanying this type of proustite damage. There is, for example, a buildup of arsenic at the crater rim and a deficiency at the center. On the other hand, the center of the crater is rich in silver relative to the rim. The sulfur distribution is asymmetric as suggested by the yellow plume as indicated above.

Other types of proustite surface damage as illustrated in the last report were explored using the technique described above and no evidence for stoichiometric changes were seen in these experiments. Thus we conclude on the basis of this brief investigation that proustite damage is accompanied by chemical decomposition only for catastrophic effects caused by continuous illumination.

An added fact in connection with obtaining the above data is that proustite is damaged by the scanning electron beam used to excite the x-ray microprobe unless the beam current is kept at a relatively low value. An example of this type of e-beam damage is shown in Fig. 3.

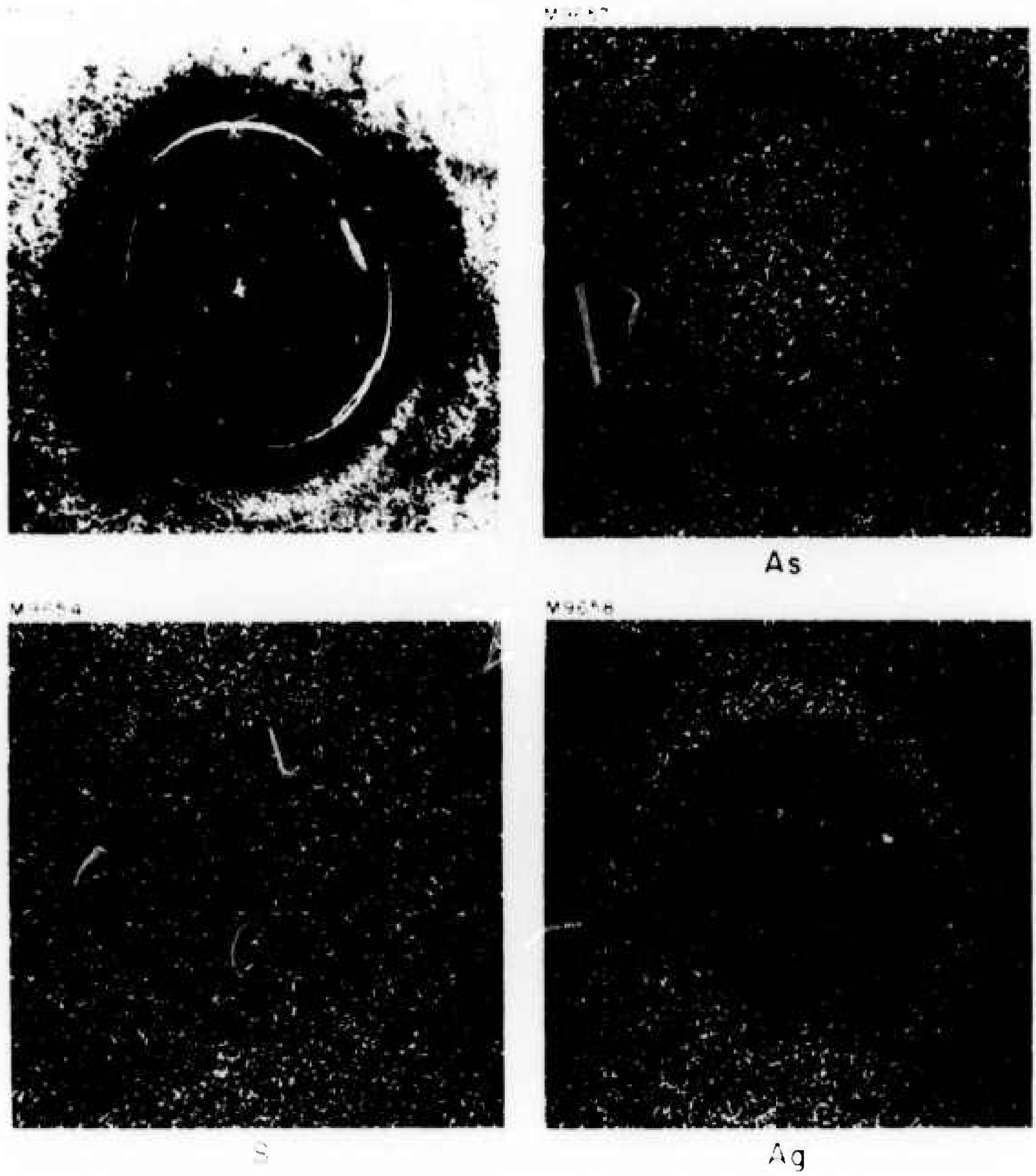

Fig. 2. Electron probe x-ray micrographs of a molten crater on proustite showing relative concentrations of Ag, As, and S.

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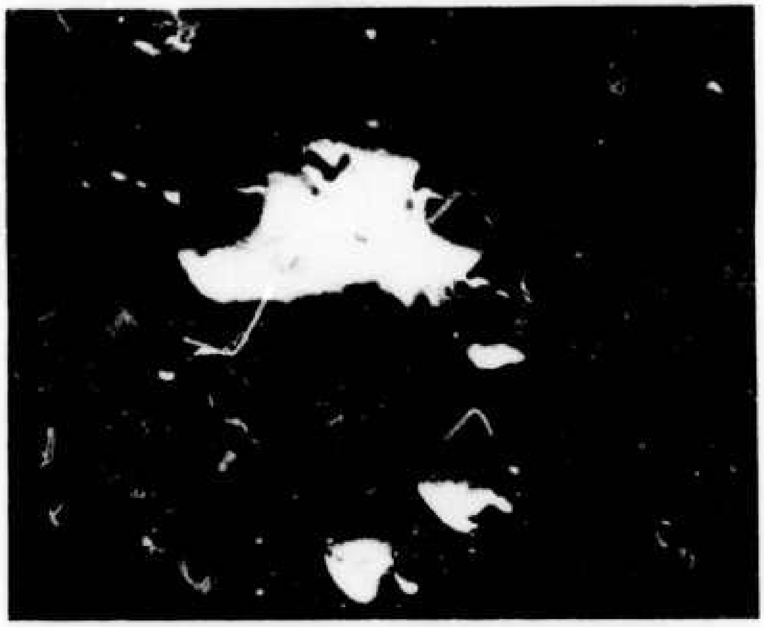

LASER DAMAGE CRATER

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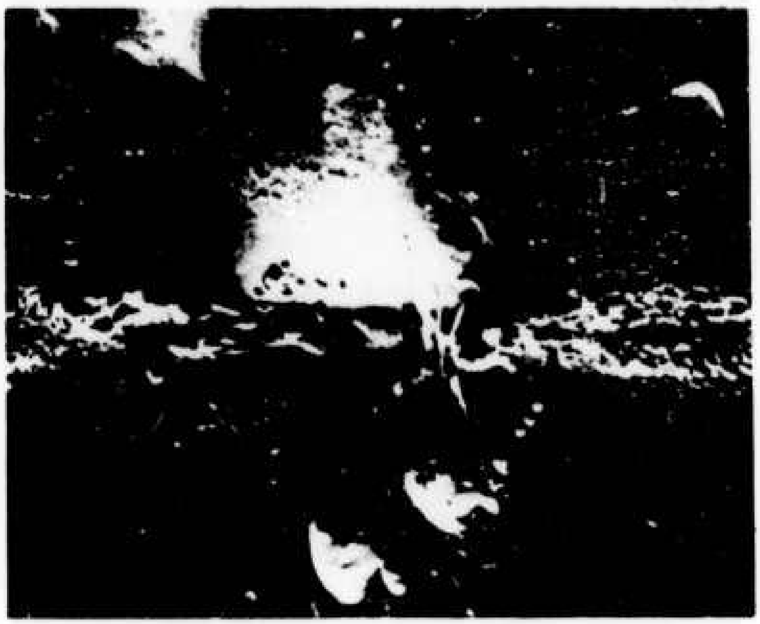

DAMAGE CRATER AFTER ELECTRON SCAN

|← 10 μm →|

Fig. 3. Scanning electron micrographs of laser damage site on proustite before and after electron microprobe scan showing damage to proustite surface due to high current electron beam.

After this damage was observed all subsequent scans were preceded and followed by the taking of scanning electron micrographs (a very low current requirement). In this way the region of interest could be examined after the microprobe analysis was carried out to ensure that a nondamaging beam current was employed.

D. Search for Impurities in Proustite

We have also used the x-ray microprobe technique for exploring for the presence of surface contaminants. As reported earlier, the near threshold damage morphology for proustite surfaces shows small molten globules clustered around surface scratches. It is possible that these scratches or grooves could collect impurities that would be difficult to remove by conventional cleaning procedures or that they might contain particles of polishing compound.

Even though the polishing abrasives used (SiO_2, Al_2O_3) do not absorb 1.06 μm light, there is the possibility that the presence of abrasive particles on the surface could give rise to some increase in local absorption in the vicinity of surface scratches.

The electron beam x-ray microprobe apparatus was used to explore for the presence of impurities on the surface. For a given sample, many areas of the surface were scanned. Typically the electron beam would illuminate a region of the surface 250 μm square and the x-ray spectrometer would scan over all the elements, the detector output being fed into a chart recorder. We have found from these measurements that except for a weak chlorine peak, there is no indication (within about 1 part in 10^3) of the presence of either aluminum or silicon.

Thus we conclude that there is no appreciable concentration of foreign impurities in the region of surface irregularities and that little or no abrasive particles are present in surface scratches or grooves.

E. Surface Damage in Lithium Niobate, $LiNbO_3$ (Ion Polishing Experiments)

During this reporting period a number of experiments have been performed on surface damage in $LiNbO_3$. The main thrust of this work has been to explore the possibility of improving the surface damage threshold by ion polishing. The status of this work at present is inconclusive. A brief summary of the results obtained so far is that no improvement in damage threshold has yet been obtained on ion polishing. However this result must be qualified by the observation that the damage seen so far in all the $LiNbO_3$ samples appears to be limited by inclusions. The quality of surface finish obtained by ion beam polishing is also strongly affected by the presence of surface inclusions.

Previous experience at HRL with ion sputtering of $LiNbO_3$ has led to a distinct sequence of steps used in the ion polishing experiments carried out during this period. Briefly described, the initial removal of material is carried out at a relatively high ion energy followed by periods of lower ion energy toward the end of the treatment. The latter stages act as a kind of annealing treatment; if they are not performed the sample is left with a substantial surface charge layer.

All ion polishing treatments carried out so far were performed in the following sequence:

3 kV for 80 min at 140 $\mu A/cm^2$
1 kV for 60 min at 140 $\mu A/cm^2$
0.5 kV for 30 min at 70 $\mu A/cm^2$

An argon ion beam was used. The beam strikes the surface at an angle of 70^o from normal and the sample is rotated during exposure to the beam.

All the samples studied were ion polished on one end only. Damage thresholds were carried out on the ion polished end and compared with measurements performed on the abrasively polished end. All damage measurements were taken with the 3.3 cm lens using the output from the Nd:YAG laser at 1.06 μm. The pulse duration was 17.5 nsec and the spot size at the sample surface was 45 μm (1/e radius for electric field).

Damage threshold power densities are presented in Table II.

We see from the data in Table II that the ion polished surfaces have typically lower damage thresholds than the abrasive polished surfaces. However, it should be pointed out that the damage appears to be limited in all cases by the presence of inclusions. We base this statement on the following:

1. In a number of cases where we attempted to generate entrance surface damage by focusing the laser appropriately we generated bulk damage instead. The bulk damage was characterized by either a series of small fractured sites along the beam or by one or two internal cracks randomly located in the material.

2. The ion polished surfaces were characterized by a number of irregularities not present on the abrasively polished surface which first appeared to be depressions or small craters but subsequently were seen to be small raised mounds. The presence of these irregularities suggests the existence of surface inclusions that have a different rate of removal in the ion beam than the host material. This type of surface feature is illustrated in the scanning electron micrographs of Fig. 4. For this surface the ion exposure was prolonged in order to emphasize the surface features.

Thus we conclude that the samples of $LiNbO_3$ studied so far have a sufficiently high density of inclusions that essentially all the damage levels measured so far are not a measure of intrinsic material. Hence the possibility of improving the material quality via ion polishing is still an open one.

(a)

(b)

Fig. 4. Scanning electron micrographs of surface features on $LiNbO_3$ after ion beam polishing.

TABLE II

Surface Damage Thresholds for $LiNbO_3$ Samples at 1.06 μm
Ion Polish versus Abrasive Polish

Sample	Threshold Power Density,[a] GW/cm^2		Number of Thresholds Measured	
	Abrasive Polish	Ion Polish	Abrasive Polished End	Ion Polished End
A	1.01	—	14	—
B	2.29	1.20	7	10
C	1.10	1.24	11	6
D	2.82	2.40	10	10
E	1.43	0.76	9	5

[a]The values are given as the total power divided by the beam area defined as πa^2 where a is the $1/e$ radius for the electric field. The on-axis power densities are twice the values listed in the table.

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III. STUDY OF TRANSMITTED, REFLECTED, AND SCATTERED LASER LIGHT DURING THE GENERATION OF SURFACE DAMAGE

During this reporting period we have pursued a series of measurements reported in Semiannual Technical Report No. 1. The purpose of this work is to characterize in detail the laser pulse with respect to instantaneous intensity, total energy, and spatial properties during the generation of surface damage.

To this end we have examined the temporal shape of both transmitted and reflected pulses as a function of incident power below and above damage threshold for both entrance and exit surfaces. The total integrated pulse energy has been monitored as a function of power for both reflected and transmitted light. Besides having examined the light that is reflected specularly from the sample surface we also measured the temporal and spatial properties of light that is scattered out of the main reflected beam at moderate angles. For the studies we have used the single mode ruby laser focused on sapphire samples, but it is evident from other cursory observations that the results apply more generally to other materials and optical wavelengths.

A. Integrated Transmission and Reflection of Laser Damage Pulses

In the last report we presented data on integrated percent transmission as a function of power for laser pulses. Further work has been carried out during this period in which we measured the reflected energy as well. The main motivation for this work has been to determine whether the decrease in transmission of damaging pulses is accompanied by a corresponding increase in reflection as might be expected if the surface plasma density reaches a sufficiently high value.

The experimental setup for monitoring the various pulses is shown in Fig. 5. For the transmission experiments, the output from the detector located after the sample (detector No. 2) is integrated electrically and displayed on one trace of a dual beam oscilloscope (Tektronix 555) while the signal from the detector that monitors the incident energy (detector No. 1) is displayed on the other trace.

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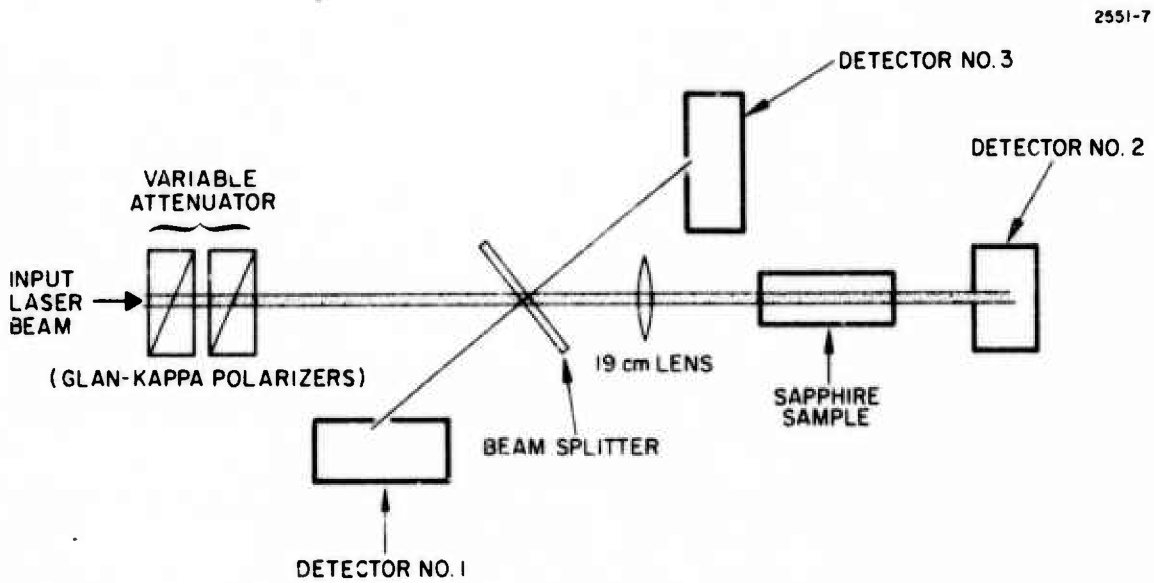

Fig. 5. Experimental setup used in transmission and surface reflection measurements.

Appropriate ratios are measured over a range of incident power from below damage threshold to many times above threshold. In a similar manner the percent reflected energy was measured in another set of experiments in which the reflected light that passes back through the focusing lens is monitored by detector No. 3 and displayed on the oscilloscope in place of the transmitted signal. The results of these measurements are shown in Fig. 6.

The percent transmission data that appeared in the last report are presented again for comparison with the reflectivity data. We see from these curves that both transmission and reflection show a monotonic decrease as the power increases above damage threshold.

B. Temporal Study of Transmitted and Reflected Damaging Pulses

A series of experiments was carried out to simultaneously study the temporal shape of transmitted and reflected pulses as a function of incident power. The purpose of these experiments was twofold: First, to determine whether the sharp drop in transmission at the time of damage formation is accompanied by a corresponding change in reflectivity at the same time, and second, to explore the possibility that some temporal irregularities might be detectable in the back reflected pulse in the absence of catastrophic damage. In essence we were also exploring for clues for precatastrophic damage information.

As we will see later, examination of the back reflected light showed no indication of precatastrophic behavior but there was some evidence for such behavior when small angle backscattering away from the directly back-reflected beam was studied.

For these experiments the outputs from detectors 2 and 3 in Fig. 5 were individually displayed on separate Tektronix 519 oscilloscopes. The use of either line filters or Wratten No. 70 red filters assured that only laser light was detected. Both oscilloscopes were externally triggered by a pulse taken from an additional biplanar photodiode that is normally used to monitor the oscillator output. This external triggering scheme ensured that the two oscilloscopes are

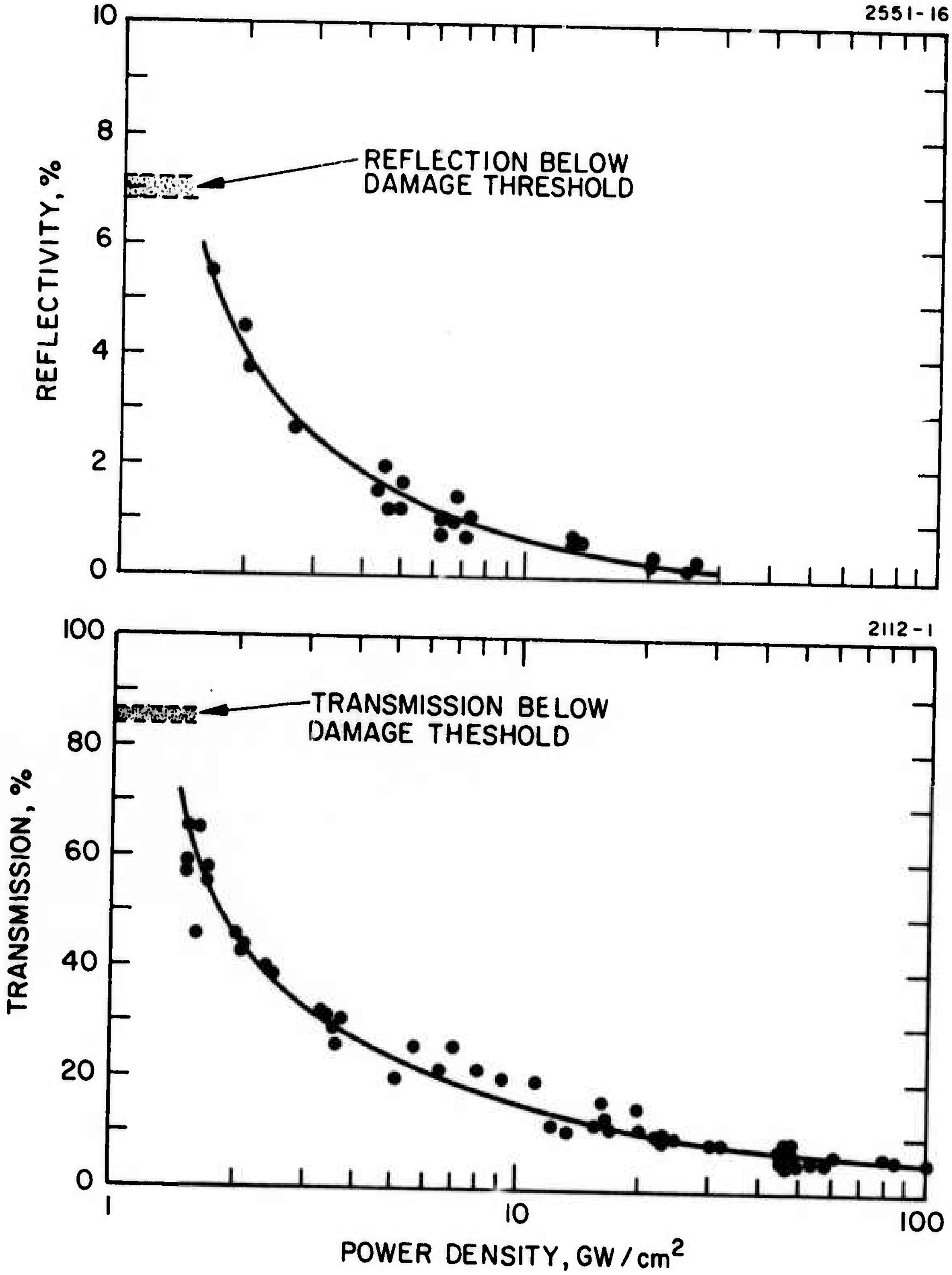

Fig. 6. Integrated transmission and reflectivity of damaging laser pulses as a function of incident power on sapphire surfaces.

triggered at the same time (within less than 1 nsec). In this way variations in pulse shape for transmitted and incident light can be temporally related even though they are separately recorded.

For each of the oscillographs recorded a double exposure was taken. In the first trace we recorded the signals when the laser was fired at the sample with the focusing lens removed from the optical path. For the second trace, the lens was appropriately positioned so that the light was focused on the surface of interest and the laser was fired again. The reproducibility of the ruby laser-amplifier combination is such that in each case (i.e., with lens and without lens) the total energy incident on the sample was the same (within ~5%). Hence, in the absence of damage, each transmitted and reflected oscilloscope photograph shows a pair of traces of the same shape and amplitude. When damage occurred the temporal variations of the damaging pulse can be compared relative to the nondamaging pulse both with respect to transmitted light and reflected light.

Experiments of this type were carried out in detail for both entrance and exit surface damage on sapphire and the results of representative shots for different incident laser power are shown in Figs. 7 and 8. Because of the difficulty in reproducing high speed oscilloscope photographs, tracings of these photographs are shown. After a great deal of difficulty it was possible to obtain reliable triggering and to eliminate all influence of stray scattered light on the recorded signals.

The following features should be pointed out concerning the traces in the figures under discussion:

- The back-reflected pulse cuts off at the same time as the transmitted pulse.
- Most temporal irregularities in the transmitted pulse are duplicated in the reflected pulse.
- The back-reflected intensity for a damaging pulse is always less than or equal to that of the reference pulse. That is, the specular reflectivity of the interface during the occurrence of damage is always less than that of the undamaged interface.

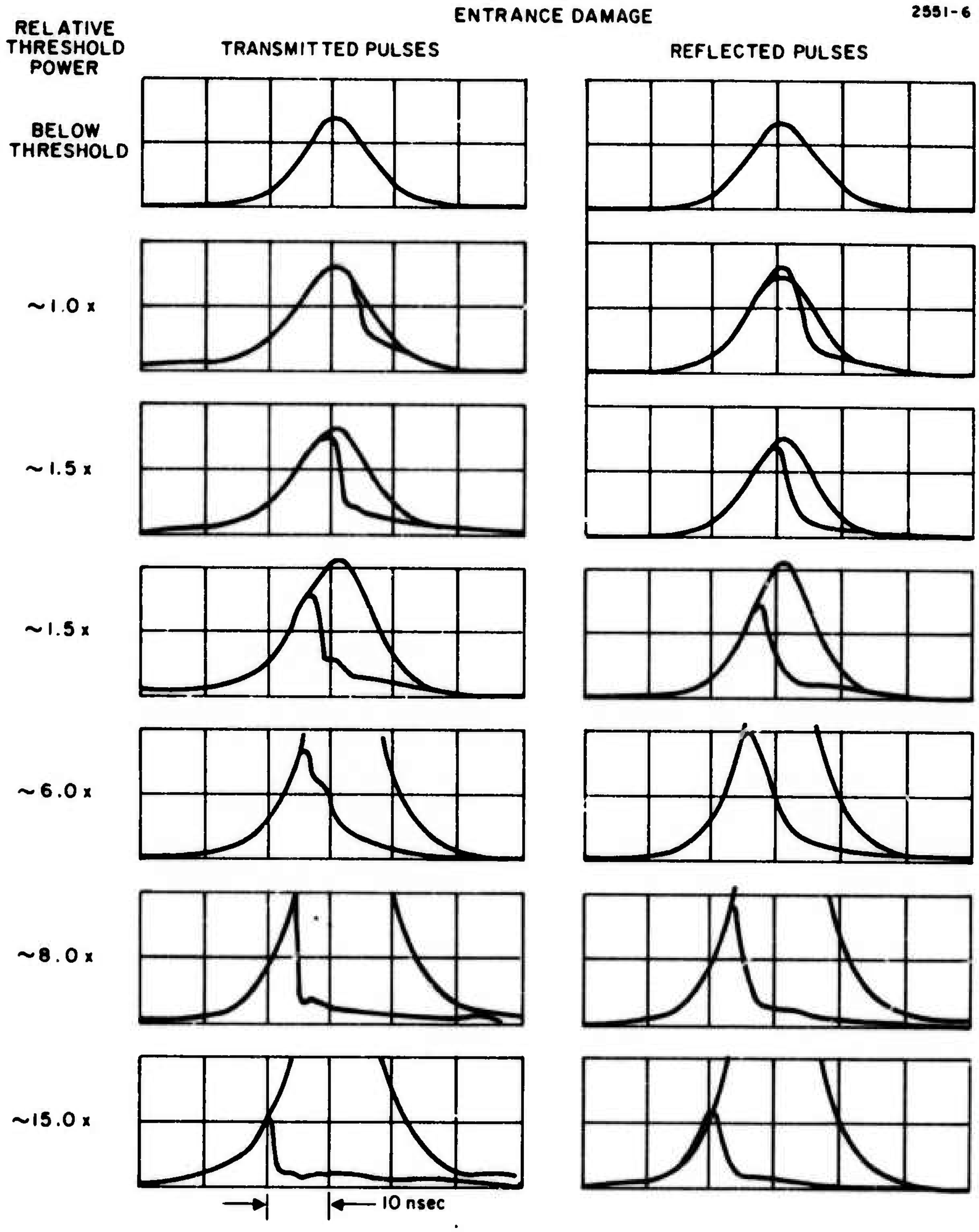

Fig. 7. Temporal shapes of transmitted and reflected pulses for different powers relative to threshold for entrance damage.

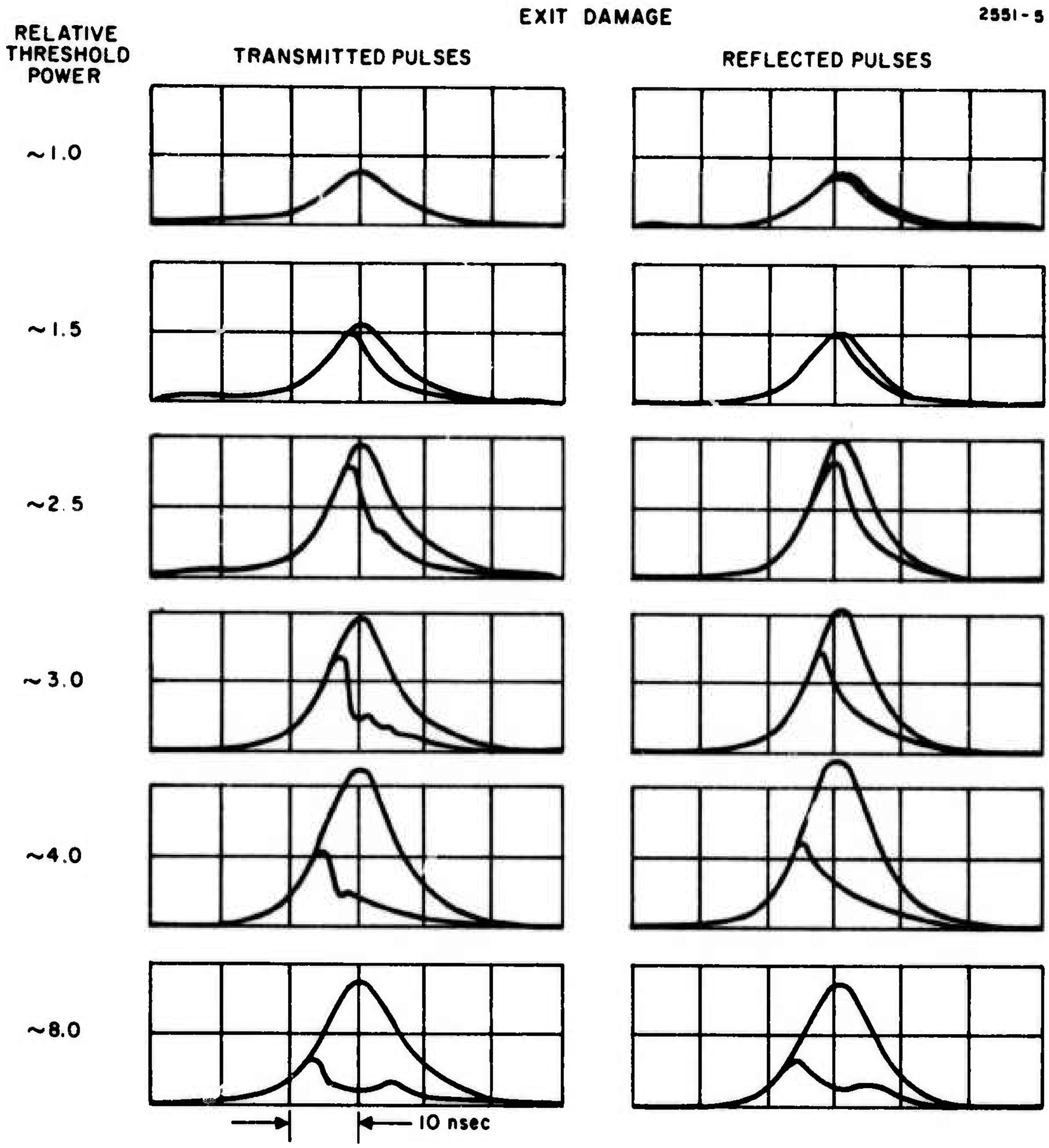

Fig. 8. Temporal shapes of transmitted and reflected pulses for different powers relative to threshold for exit damage.

Because of the lens diameter (1 in.) and distance from the sample surface (20.7 cm) the total amount of reflected light collected in the above described experiments would have spanned a maximum half angle of 3.5°.

C. Measurements of Backscattered Laser Light From the Surface During Damage

Since both the transmitted and reflected pulses show a sharp decrease in intensity following the creation of damage one might be inclined to attribute the loss of light to absorption by the damaged region and/or surface plasma. However, there is also the possibility that an appreciable amount of the laser light is scattered out of the forward and back direction as well as absorbed.

A measurement of the amount of scattered light would require the means for collecting and measuring the light scattered over all directions or a knowledge of the scattering distribution and subsequent sampling of the light over a particular solid angle. Measurements of this type were not carried out in our experiments but a few experiments were conducted that were aimed at determining the temporal behavior of scattered light. Again, the purposes were: (1) To explore the possibility that an appreciable amount of light might be scattered out of the main beam, (2) to obtain a temporal correlation between the scattered light and transmitted or specularly reflected light, and (3) to explore the possibility that some indication of precatastrophic damage might be found in the temporal characteristics of the scattered light.

The experimental setup used for these experiments is shown in Fig. 9. It is similar to the previous setup except that a white card (3 in. square) with an aperture was placed approximately halfway between the focusing lens and the sample. Detector No. 3 was placed to monitor the diffuse laser light scattered back from the sample surface onto the white card. The aperture diameter (6.5 mm) was substantially larger than the convergent beam diameter (1.7 mm) at the point where it passed through the hole in the card.

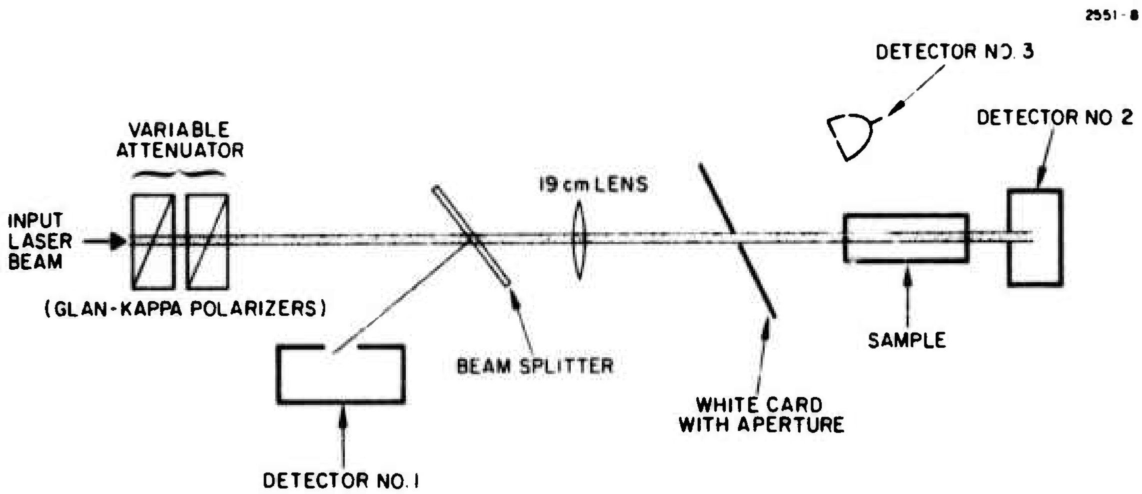

Fig. 9. Experimental setup used in backscattering measurements.

The experimental data were obtained in a similar way that the previously described data were taken; the laser was fired once with the lens removed and the second time with the lens placed appropriately in the beam, both pulses being recorded on the same oscilloscope film.

The results of a number of these experiments are illustrated in Fig. 10. Here the data are displayed differently than before in that the scattered pulse has been inverted relative to the transmitted pulse so that the temporal relationship can be more easily seen.

The general qualitative features seen in these traces are:

- The scattered light for damaging pulses is characterized by a sharply rising spike followed by a more slowly decaying tail.
- The peak of the scattered spike occurs at the same time that the transmitted pulse begins its sharp cutoff.
- The scattered intensity begins to rise from its background level before the transmitted pulse shows any appreciable irregularity.
- In cases where very slight damage occurs so that the transmitted pulse shows little or no temporal irregularity, there is still an appreciable temporal irregularity in the scattered light.

The sensitivity of the backscattered amplitude shown in the above experiments indicates that this type of monitoring is a more sensitive indication of surface damage than examination of either the transmitted or the specularly reflected light. Moreover, the fact that the backscattered light begins to rise in intensity relative to the background before the transmitted light shows signs of cutoff suggests that some evidence for precatastrophic damage may be indicated from this kind of measurement.

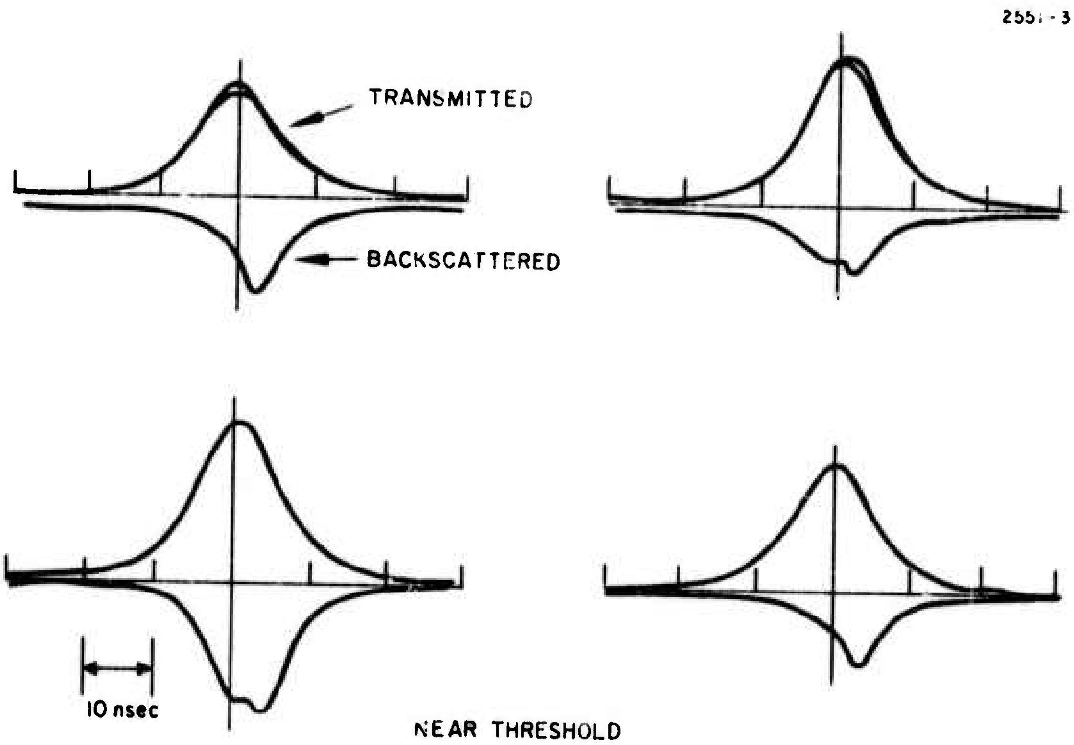

Fig. 10(a). Temporal profiles of laser pulses transmitted through and backscattered by sapphire samples at different powers relative to entrance surface damage threshold.

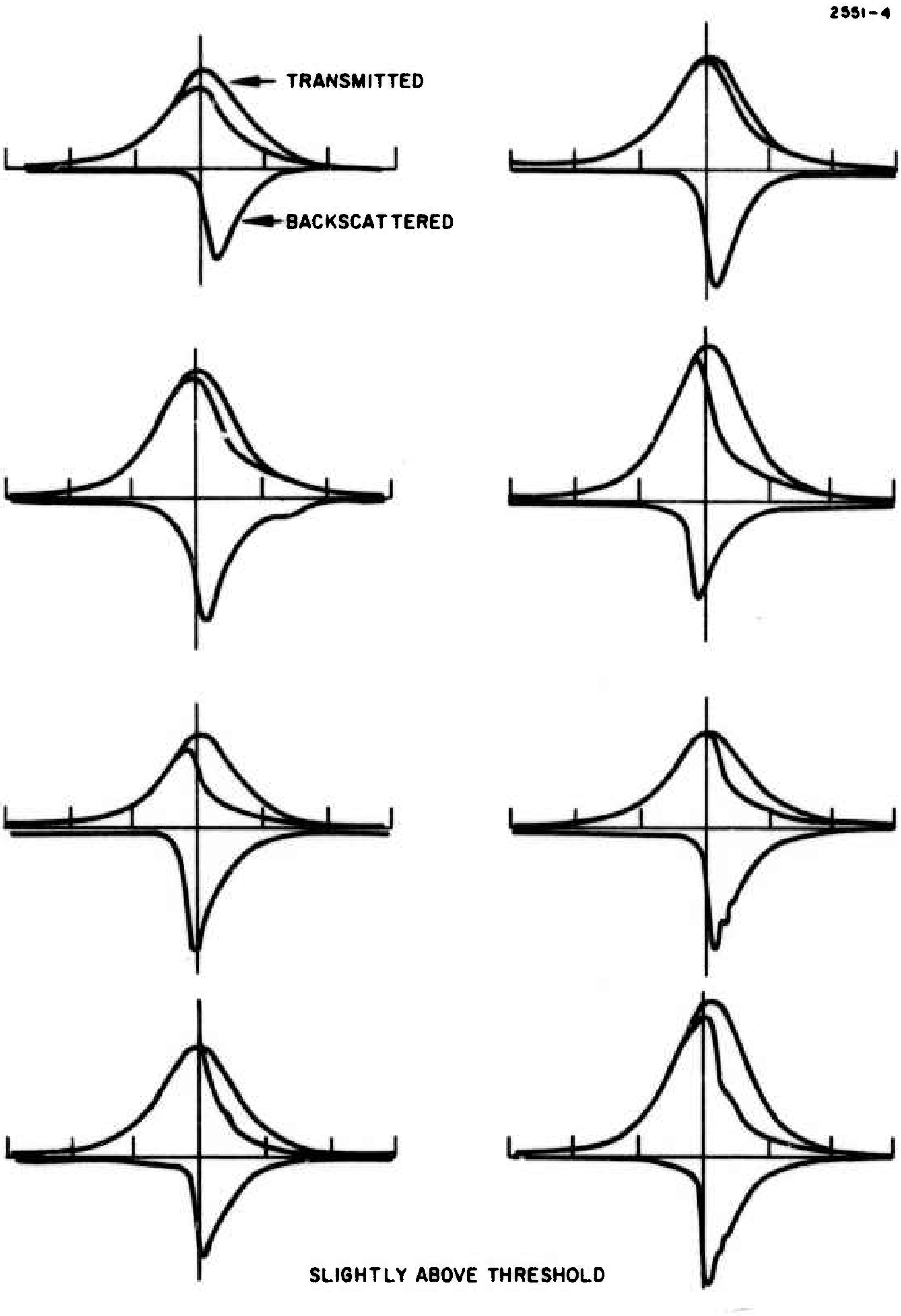

Fig. 10(b). Continued.

D. Precatastrophic Damage Experiments

Tentative evidence for precatastrophic damage indications were seen in a few instances in the absence of damage where the scattered light showed a definite irregularity compared with the background pulse. In these cases the transmitted pulses were essentially identical with or without the focusing lens in place whereas the backscattered pulse with the lens in place had a different shape than the pulse observed with the lens removed.

Examples of this behavior are shown in Fig. 11. We see that the scattered pulse with the lens in place is different in shape from the one where the lens is removed. In these examples the laser power incident on the surface was very close to the value where damage occurred but not sufficiently large to produce any detectable damage on that particular shot. All the pulses in the examples shown were followed by damage producing pulses of slightly higher energy.

Because of the detector sensitivity it is difficult to obtain large signals at these levels of illumination. More detailed experiments of this type with higher signal levels will indicate whether the effect observed is really an indication of precatastrophic damage.

Since the background pulse in these experiments is one in which the lens is removed from the system, the diameter of the beam is passing through the 6.5 mm aperture is different in the two cases. (The diameter of the focused beam at the $1/e^2$ intensity points is 1.7 mm and that for the unfocused beam is ~4 mm.) Also, the size of the spot striking the sample surface is substantially different. Since the general background of scatter on the card will depend on the surface smoothness and general distribution of irregularities, scratches, etc., the level of the scattered signal is not expected to be the same for the two pulses.

The temporal shapes however, should be the same for the scattered pulses as for the transmitted pulses in the absence of damage unless some subtle phenomenon is taking place.

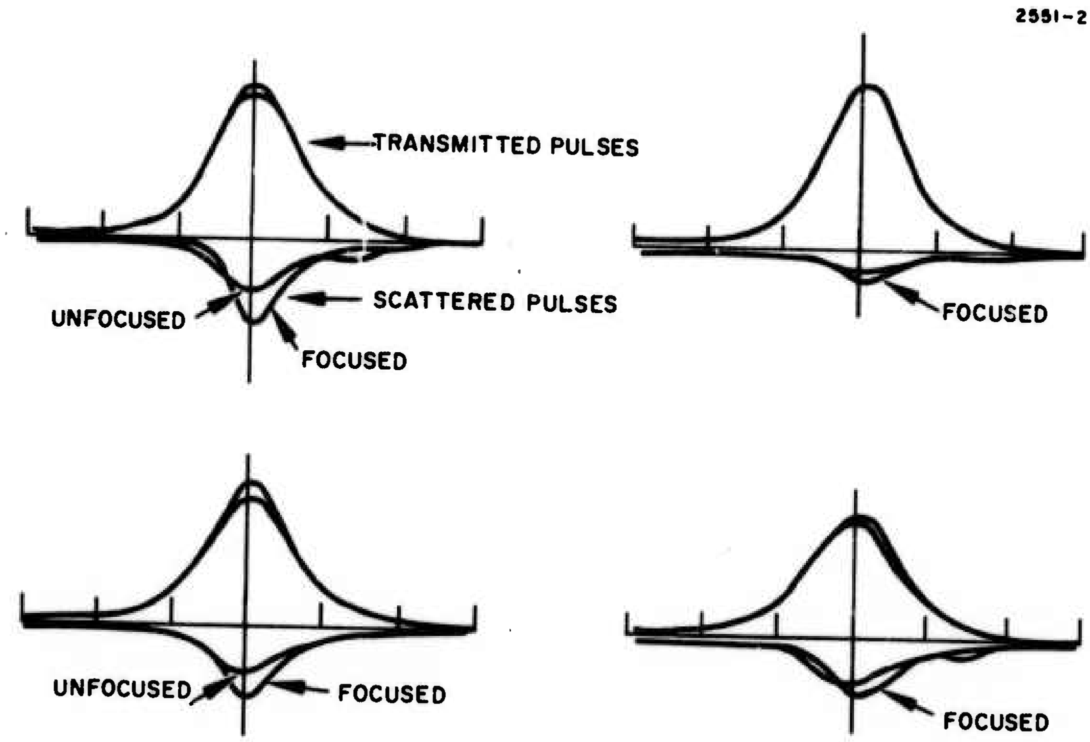

Fig. 11. Backscattered pulses compared with transmitted pulses below damage threshold showing temporal irregularities in the backscattered pulse.

A possible explanation is a precatastrophic surface phenomenon such as a slight local heating resulting in an effective local curvature at the surface that could deflect the beam temporarily off axis. Another is the production of a microscopic plasma that is not visually detectable but that results in a subtle off axis backscattered component.

At this time we cannot preclude the possibility that the phenomena illustrated in Fig. 11 might arise from a spatial peculiarity of the focused beam relative to the unfocused beam that could result in a difference in temporal behavior of the scattered light. Because some of the light detected occurs in the wings of the spatial distribution, there may be a subtle temporal difference between the two beams, especially if the laser is not oscillating in a single longitudinal mode. This possibility is not a likely one, but only more detailed experiments will allow us to eliminate it.

E. Spatial Properties of Backscattered Light From Damaging Pulses

The fact that a measurable amount of light is scattered out of the specularly reflected beam for damaging pulses led to the question concerning its spatial distribution. To this end an experiment was carried out, as illustrated in Fig. 12, where the specularly reflected light was allowed to strike a photographic film (Polaroid Type 47). In addition to photographing the reflected beam as a function of incident total energy, the spot was photographed for different diameter apertures placed in the beam between the lens and the sample. In this way angular information could be obtained for some of the off-axis features.

Qualitative features of these beam profiles are shown in Figs. 13 and 14. At low powers a smooth circular spot is seen that remains essentially unchanged when damage threshold is reached and even substantially above damage threshold (~2x). At higher powers (above twice threshold) signs of off-axis features begin to appear and an increasing amount of light is detected in the form of irregular rings and bright spots.

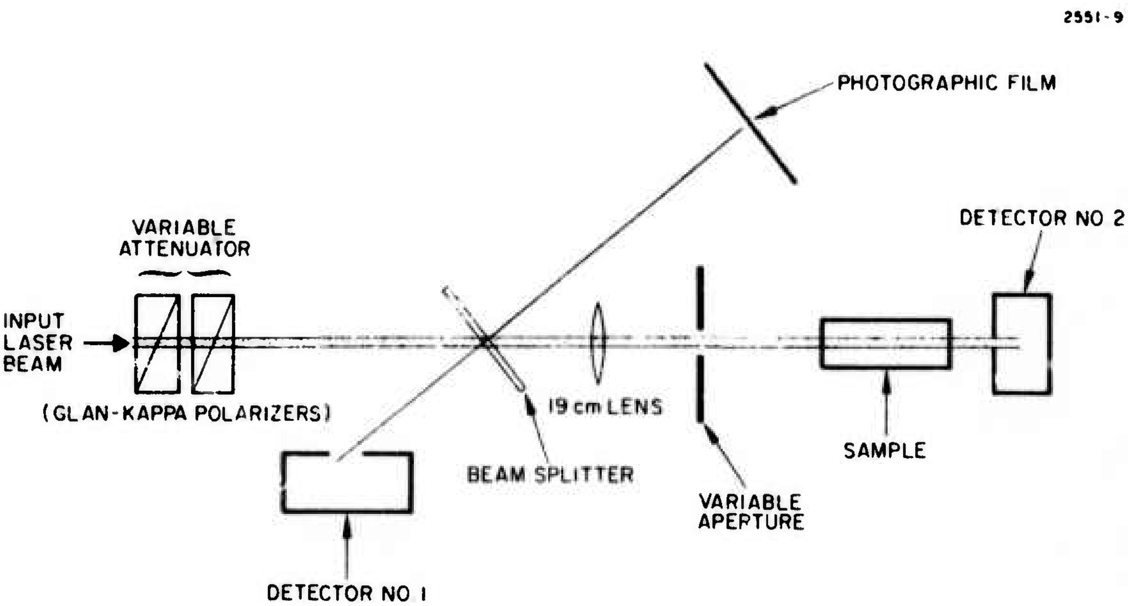

Fig. 12. Experimental setup for photographing spatial features of specularly reflected light.

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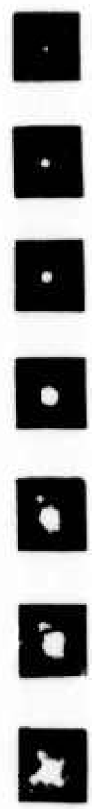

Fig. 13.
Photographs of back-reflected beam for increasing laser power above damage threshold (from 1 to 10x).

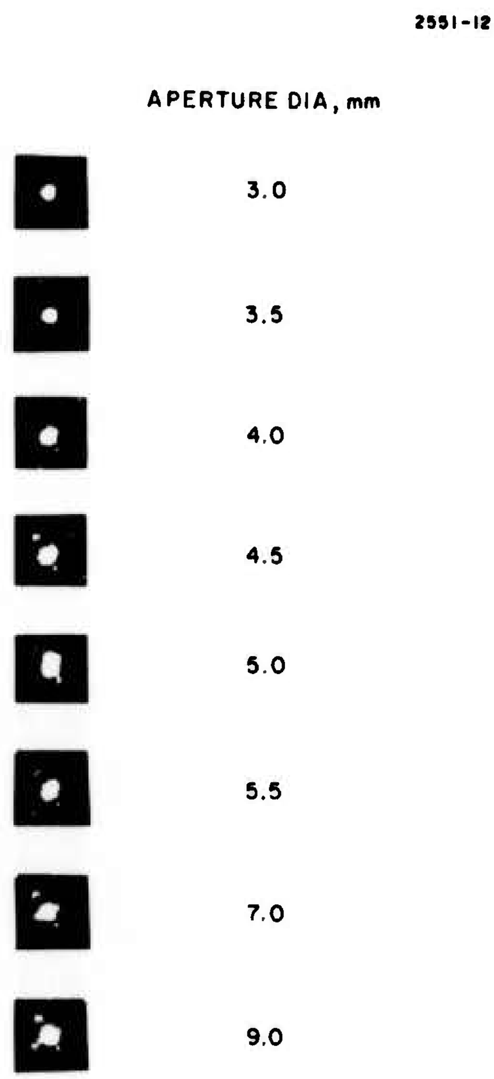

Fig. 14.
Photographs of back-reflected beam for different aperture sizes between lens and sample. Constant laser power for each shot ~10x threshold.

The angular extent over which the pattern is detected was measured by varying the size of the aperture between the lens and the sample. It was found that an aperture diameter between 5.5 and 7 mm was the smallest that could be used to give the same pattern as obtained with no aperture in the system (Fig. 14). (The effective limiting aperture caused by the lens would be 12 mm at that position.) Hence the off-axis features in the photographs extend over a half-angle of from 1.5^{o} to 2^{o}.

It should be pointed out here that the measurements of "specularly" reflected light discussed earlier included the light that is shown in the photographs of Figs. 13 and 14. Because of the setup used in those experiments, the angular acceptance of the detected light was on the order of 3^{o} half-angle. Hence, even though a detectable amount of light is scattered out of the specular beam, the total amount of light collected over the 3^{o} half angle shows the general decrease with power above damage threshold.

Along the same line of discussion, the temporal spikes detected in the off-axis scattering experiments (Fig. 10) were seen to occur at half-angles larger than 2^{o}; none of the light detected in those experiments was of sufficient intensity to be recorded on the beam photographs.

F. Summary of Pulse Temporal and Spatial Experiments

The results of the above measurements can be summarized as follows:

1. The damaging pulse shows a sharp temporal cutoff both in transmission and reflection for light collected over a cone of 3.5^{o} half angle.

2. Photographs of the back reflected light show that a considerable amount of this light is scattered out of the main beam at small angles (less than 2^{o}) in the form of bright spots and diffuse rings.

3. In addition to the small angle backscatter seen in the photographs, an appreciable amount of light is scattered back at larger angles that is not seen photographically.

4. The light scattered back at larger angles (within a 23° half-angle cone) shows a sharp temporal spike at the time the transmitted light and specularly reflected light cuts off.

5. Tentative evidence indicates the possibility that precatastrophic effects can be explored through observation of this large angle backscattered component.

6. Although it is almost certain that the damaged surface site and associated plasmas absorb a measurable amount of the incident pulse, it is not possible to obtain an accurate measure of the light absorbed during damage without integrating all the scattered light.

IV. PLANS FOR NEXT PERIOD

The precatastrophic damage evidence obtained in preliminary results reported here will be explored further during the next period in an attempt to obtain more substantial data. The conditions under which this phenomenon is observable will be explored and detailed surface characterization will be employed. Ion polishing experiments on $LiNbO_3$ and other materials of interest will continue and surface damage threshold will be evaluated under different conditions of ion exposure. The spot size dependence of proustite surface damage will be explored further in an attempt to check the results obtained so far. Also, if possible the Nd:YAG laser will be modified so that larger pulse durations can be studied (~50 nsec) and compared with the results obtained so far.